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Near-Global Survey of Cloud Column Susceptibilities Using ISCCP Data

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Abstract. A new parameter, cloud *column* susceptibility, is introduced to study the aerosol indirect effect, which describes the aerosol indirect effect more directly without assuming how cloud droplet size will respond to changes of droplet number concentration. Between the two approaches that used to retrieve cloud column susceptibilities, the one that makes no assumption of constant liquid water content leads to smaller, even negative cloud column susceptibilities. This finding is consistent with results of model studies and observations from the 1998-1999 Indian Ocean Experiment (INDOEX) that suggest that cloud liquid water content may be reduced during aerosol-cloud interactions. The results of this survey suggest that using constant liquid water content in models may lead to significant overestimation of the aerosol indirect effect.

1. Introduction

Among possible radiative forcings that can cause long-term climate change, the effect of changing tropospheric aerosols on cloud properties is the most uncertain (0 to -1.5 Wm^{-2}). A recent model study further suggests that the indirect aerosol effect may play vital role in global climate change [*Hansen et al.* 1997].

Twomey [1991] defined the cloud susceptibility as cloud albedo change versus number concentration change of cloud droplets, dα/dN, which indicates where clouds are more susceptible to the cloud-aerosol interaction. However, the accuracy of calculation and feasibility of remote sensing of this parameter have been limited by the assumptions used, i.e., cloud liquid water content and geometrical thickness were held constant [*Platnick and Twomey,* 1994, *Taylor and Mchaffie*, 1994]. These assumptions are often violated even for non-precipitating clouds. Observations show different cases where liquid water content increases, decreases, or approximately holds constant [see *Han et al.* 1998a and references therein]. The cloud geometrical thickness also changes during the aerosol-cloud interactions [e.g., *Hobbs et al.* 1970].

When cloud liquid water content and geometrical thickness are held constant, the cloud susceptibility is in the form

$$\frac{d\alpha}{dN} \approx \frac{\partial\alpha}{\partial\tau}\frac{d\tau}{dN} \approx \frac{\partial\alpha}{\partial\tau}\frac{\tau}{3N} = \frac{4\pi\rho_w}{9w}\alpha(1-\alpha)r_v^3;\ \alpha \approx \frac{(1-g)\tau}{2+(1-g)\tau} \quad (1)$$

where w represents the cloud liquid water content and was assumed to be 0.3g/m^3 in *Platnick and Twomey* [1994]. This form is valid only for cases when the liquid water content is approximately constant and its value can be obtained from other measurements. However, in general, cloud liquid water content varies strongly ranging from 0.2 g/m^3 up to 5g/m^3 [e.g., *Pruppacher and Klett,* 1997, p23], which introduces more than one order of magnitude uncertainty in cloud susceptibility if global survey of this parameter is conducted.

To avoid the difficulties caused by variations of liquid water content, we introduce a new parameter, *cloud column susceptibility*, which has several advantages. First, it can be easily used in model calculations without the assumption of constant liquid water content and geometrical thickness. Second, it can be retrieved globally using satellite data without the assumption of an average value of liquid water content. Third, this new parameter includes the effect of variations of cloud geometrical thickness during cloud-aerosol interactions. The new parameter describes the indirect aerosol effect more directly without assuming how cloud droplet size will respond to changes of number concentration.

2. Method

The cloud column susceptibility is defined by

$$S_c = d\alpha / dN_c \quad (2)$$

where α is cloud spherical albedo and N_c is cloud column droplet concentration. The column droplet concentration is defined by

$$N_c = \int_0^h n(h')dh' \quad (3)$$

where $n(h')$ is the profile of cloud droplet number concentration and h is the cloud thickness. The retrieval of N_c, validation, and results of a global-survey were described by *Han et al.* [1998b].

The cloud column susceptibility is retrieved using two approaches, i.e., with and without the assumption of constant liquid water content. In both approaches, cloud optical thickness and effective droplet radius are retrieved for each pixel using the three-channel method [*Han et . al.* 1994]. Pixel sizes are about 5 km x 1 km and have been sampled at intervals of about 30 km.

2.1. Approach One: Assuming Constant Liquid Water Content

Using constant cloud liquid water content and thickness, the cloud column susceptibility can be expressed as

$$S_c = \frac{d\alpha}{dN_c} \approx \frac{\partial\alpha}{\partial\tau}\frac{d\tau}{dN_c} \approx \frac{\partial\alpha}{\partial\tau}\frac{\tau}{3N_c} = \frac{\alpha(1-\alpha)}{3N_c} \quad (4)$$

The column droplet concentration, N_c, and the spherical albedo of cloud, α, are retrieved from satellite radiance data [*Han et al.* 1998a, b]. The column susceptibility is derived at pixel level. This approach is similar to the one used by *Platnick and Twomey* [1994] in retrieving cloud susceptibility. The major difference is that the parameter retrieved here is cloud *column* susceptibility in which no assumption about the value of liquid water content and thickness are required. Hence, it can be applied in any region no matter what the value of cloud liquid water content might be.

Clearly, the calculated cloud column susceptibility by this approach will be incorrect if the assumption of constant liquid water content is invalid. In fact, the assumption of constant

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liquid water content is an additional constraint on the response of droplet size to the change of droplet number concentration,

$$\frac{dr_e}{dN} = -\frac{r_e}{3N} \tag{5}$$

2.2. Approach Two: Regression Method

In reality, cloud liquid water content has been observed to increase, decrease or remain constant. Different behaviors of liquid water content lead to different results for cloud susceptibility. To determine these behaviors for a specific cloud pixel from satellite data is very difficult. Therefore, the regression method is chosen as the second approach.

In this approach, no assumption about liquid water content is made and biases caused by this assumption are avoided. The cloud column susceptibility

$$S_c = \frac{d\alpha}{dN_c} \approx \frac{\Delta\alpha}{\Delta N_c} \tag{6}$$

is derived from statistical regression with N_c and α retrieved from satellite radiance data [*Han et al.* 1998a]. The column susceptibility values of each 2.5°x2.5° grid box are derived from the slope of a linear regression of all water cloud pixels (determined by cloud top temperature >273 K) within this grid box using one month of data. Typical pixel numbers are >100 for each grid box. If pixel number in a grid box is less than 10, no regression is performed and the grid box is left blank.

3. Results

Figure 1 is the retrieved column susceptibility for thin ($\tau \leq 15$) clouds by the first (Fig. 1a) and the second (Fig. 1b) approaches, respectively. They both show striking contrasts of cloud column susceptibilities between continental and maritime clouds expect for South America and Central Africa where the effect of rain forests is apparent. For clouds over most continents, the column cloud susceptibility is around zero ($\leq 1.8 \times 10^{-8}$ cm^2), suggesting little cloud albedo change due to the cloud column droplet concentration change. For most oceanic clouds, the column susceptibilities are high, suggesting a large albedo response to a change in the column droplet concentration. In a clean oceanic environment, for a typical maritime cloud with 300 m physical thickness, the column cloud susceptibility of 6.6×10^{-8} cm^2 means that an increase in volume cloud droplet concentration of 10 cm^{-3} would increase cloud albedo by 2.0%. This is reasonable since the volume cloud droplet concentration of a typical marine cloud is only about 40 cm^{-3}.

Case studies of cloud susceptibility from the California and Namibia marine stratus regions were described by *Platnick and Twomey* [1994] and *Taylor and McHaffie* [1994]. The results of *Platnick and Twomey*, using AVHRR for ship-track region in stratocumulus west of Washington State on March 2, 1990,

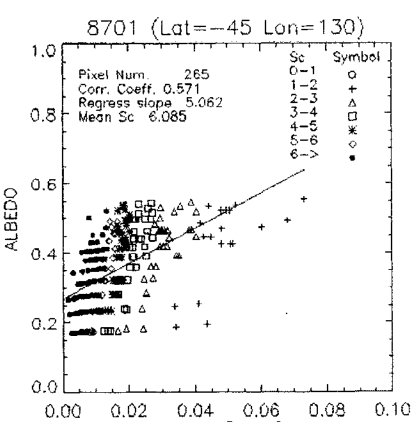

Figure 1. Cloud column susceptibility retrieved from NOAA-9 data of 1987 (a) with the assumption of constant liquid water content, (b) without the assumption of constant liquid water

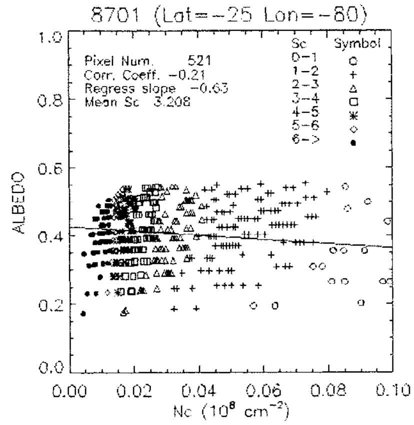

Figure 2. Example of positive column susceptibility within a grid box at south of Australia.

are used here for comparison because detailed information of cloud properties were reported. All 28 retrieved results of cloud susceptibilities in that paper are converted to cloud column susceptibilities by the liquid water path and liquid water content reported in their studies. The converted average cloud column is $10.6 \pm 9.6 \times 10^{-8}$ cm^2. The value is $21.5 \pm 8.5 \times 10^{-8}$ cm^2 if only out-of-track regions (10 cases) are considered and $4.6 \pm 1.3 \times 10^{-8}$ cm^2 if only in-track regions (18 cases) are included. The total average value is in good agreement with our results west of Washington State in April 1987 (10-12 $\times 10^{-8}$ cm^2) if constant liquid water content is assumed (Fig. 1a). Another dataset used for comparison is from observations in marine stratocumulus clouds off the west coast of southern California in the vicinity of 32°N and 120°W in July 1987 [*Radke et al.*, 1989]. According to the cloud properties supplied in the paper and using equation (1), the range of the cloud column susceptibility is from 1.5×10^{-8} cm^2 to 3.9×10^{-8} cm^2. This agrees well with the value of 2.0×10^{-8} to 4.0×10^{-8} cm^2 from Fig. 1a for July 1987 off the west coast of California. The good agreements with case studies only suggest that if the assumption of constant liquid water content is valid, the calculated cloud column susceptibilities are similar from satellite retrievals and by in situ measurements. This essentially indicates that values of τ and r_e retrieved by satellite data agree reasonably well with those obtained by aircraft measurements. However, if w cannot be held constant, both calculated cloud susceptibilities would be incorrect.

The main difference between results from Figs. 1a and 1b is that the column susceptibility is smaller if water content is not assumed to be constant in the retrieval. This difference is more significant over continents and the nearby ocean areas. For example, in July, in the West Atlantic, the average cloud column susceptibility drops from about 9.0×10^{-8} cm^2 (assuming constant liquid water content) to about 1.6×10^{-8} cm^2 (no assumption about liquid water content). This may be caused by the air pollution from the eastern United States.

The statistical susceptibility shown in Figure 1b is negative in some locations. However, the derivative $d\alpha/dN_c$ is usually thought to be positive in all cases. The reason is usually stated in terms of relation between cloud optical thickness (albedo), water content, w, and droplet effective radius, r_e:

$$\tau = \pi \int_0^h \int_0^\infty Q_{ext} r^2 n(r) dr dh' = Q_{ext} \frac{\pi \bar{r}^3 N}{r_e} h \approx \frac{3}{2} \frac{w}{r_e} h \tag{7}$$

where $\bar{r}$ is the volume average droplet radius and N the total droplet number concentration. Thus, the argument goes, when N increases, the droplet radius decreases and albedo increases, but this is true only if w is constant. If instead of this assumption, we substitute into (7) the relation

$$w = k r_e^3 \rho_w N$$

where $k=4\pi(1-b)(1-2b)$ and b ranges from 0.10 to 0.20 [*Han et al.* 1994], we have

$$\tau = \frac{3}{2}\frac{w}{r_e}h = \frac{3}{2}kr_e^2\rho_w N_c \quad (8)$$

Now, instead of an inverse relationship between τ and r_e, suggested when w is held constant, changes in τ and r_e could be positively correlated if N_c is nearly constant as indicated by some observations [*Lohmann et al.* 1999]. In fact, observations show that a decrease of cloud droplet size is more often than not linked with a decrease of the optical thickness for most clouds on the earth [*Han et al.* 1998a]. In the study of cloud-aerosol interactions, neither $\Delta w=0$ nor $\Delta N_c=0$ should be assumed *a priori*. From equation (8), the change in cloud optical thickness (hence albedo) due to changes in N_c can be written more generally as

$$\Delta\tau / \Delta N_c = 3k\rho_w r_e N_c \left[\frac{r_e}{2N_c} + \frac{\Delta r_e}{\Delta N_c}\right] \quad (9)$$

Thus, cloud susceptibility can be negative if

$$\frac{\Delta r_e}{\Delta N} < -\frac{r_e}{2N} \quad (10)$$

This condition suggests a decrease in cloud liquid water content during aerosol-cloud interactions. As will be discussed later, this is supported by results from model studies and observations.

For thick clouds ($\tau>15$), as expected, the cloud column susceptibility is mostly close to zero (not shown), which implies that a change in the column droplet number concentration has little impact on the cloud albedo.

Figure 2 is a scatter plot of data in a grid box with positive column susceptibility in January 1987 (located south of Australia). Increasing cloud albedo is roughly determined by increases of the optical thickness. Within small intervals of optical thickness, droplet sizes become smaller when the column number concentration increases, as is readily seen from equation (8) for constant τ. It shows that in this region, the column number concentrations of most thin clouds are less than 2.0 x 10^6 cm^{-2} while most thick clouds are between 1.5 to 3.0 x10^6 cm^{-2}, which makes the column susceptibility positive. A comparison between the two different approaches is also shown in Fig. 2 with the column susceptibility

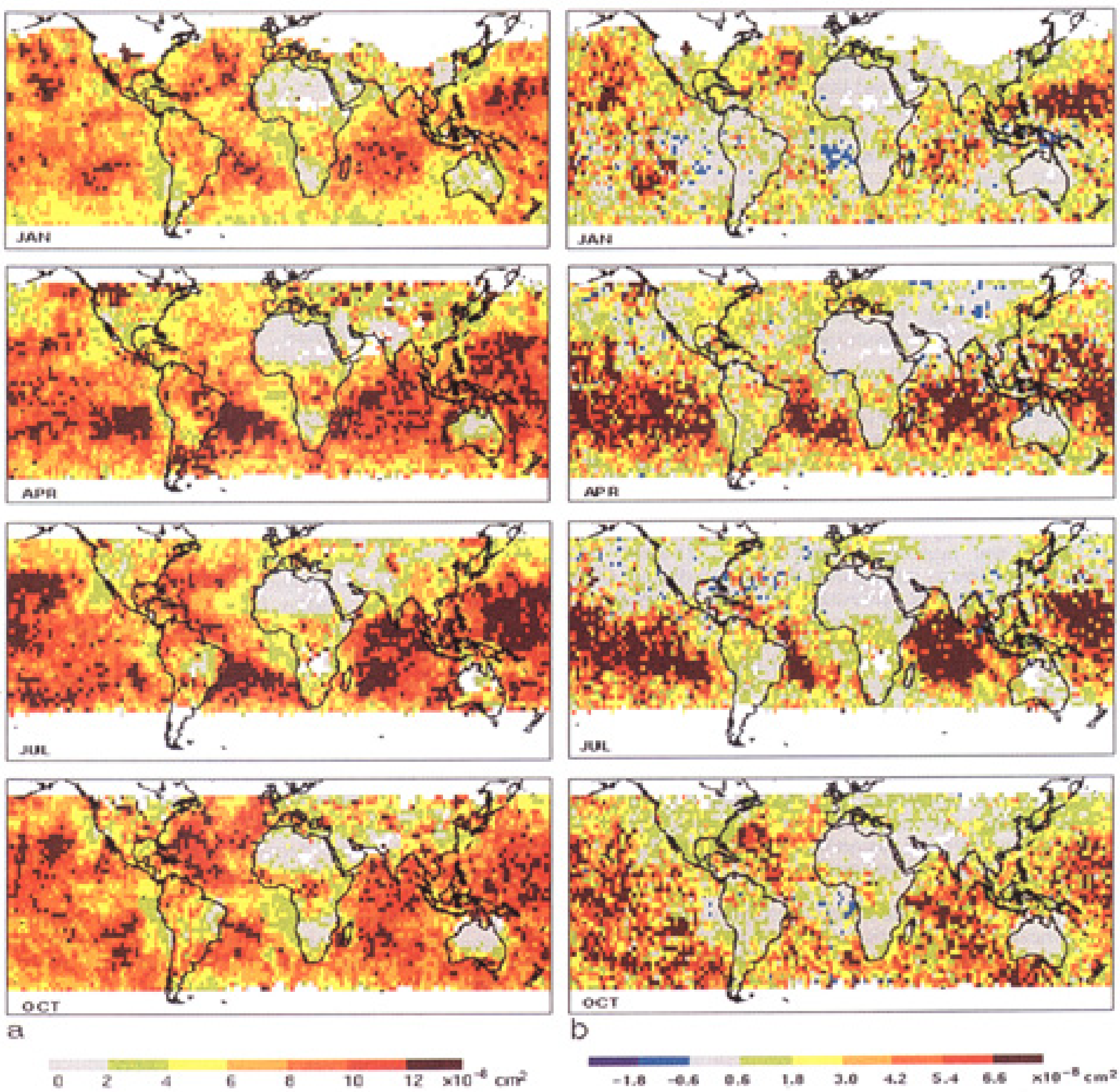

Figure 3. Example of negative column susceptibility within a grid box at the west coast of Peru.

retrieved by approach one shown in different symbols. The unit of the column susceptibility (S_c) in the figure is 10^{-8} cm^2. It is readily seen that the column susceptibility by approach one is strongly dependent on the value of N_c, which is understandable because the range of ΔN_c is much larger than the range of $\Delta[\alpha(1-\alpha)]$ in the equation (5). The average column susceptibility of all pixels from approach one is 6.1 x10^{-8} cm^2, larger than the result of 5.1x10^{-8} cm^2 by the regression method. In other grid boxes, we have seen much larger differences between results from these two approaches. This comparison suggests that holding liquid water content constant overestimates the indirect aerosol effects.

Figure 3 shows an example of negative column susceptibility during the same month (located near the west coast of Peru). The averaged column susceptibility using approach one is 3.2x10^{-8} cm^2, while the approach two yields S_c=-0.63x10^{-8} cm^2. In comparison with Fig. 2, column number concentration is larger and droplet size is smaller for all optical thicknesses. This negative S_c cannot be understood if liquid water content is held constant. However, it is consistent with model studies and observations. Model studies show that cloud top warming or cloud base cooling can lead to reduced boundary-layer mixing, which restricts the supply of water vapor and results in a thinning of cloud layers [e.g., *Lilly*, 1968; *Bougeault*, 1985; *Turton and Nicholls*, 1987). When more CCNs are activated into cloud droplets the total droplet surface area, and thus evaporation, increases due to a greater droplet concentration and smaller average size of the droplets. The increased evaporation at cloud base leads to a greater decoupling between the cloud and the subcloud layers that causes a reduction of cloud liquid water [*Ackerman et al.* 1995]. Another mechanism may lead to desiccation of clouds due to aerosol-cloud interaction is by enhanced solar absorption [e.g., *Ackerman and Toon*, 1996]. Calculations based on results from the observation during the 1998-1999 Indian Ocean Experiment (INDOEX) show that fractional cloud coverage might be reduced by 25% to 40% due to heating by aerosol absorptions [*Schwartz and Buseck*, 2000].

4. Discussion and Conclusions

A new parameter, cloud column susceptibility, is introduced that can be retrieved using satellite data and can be used to compare with model studies. Two different approaches, with and without the assumption of constant liquid water content in the cloud-aerosol interaction, were used to retrieve the cloud column susceptibility. The results from both approaches show continental and maritime contrast that is consistent with differences of aerosol loading over these two surfaces. Cloud column susceptibilities retrieved without the assumption of constant liquid water content are generally smaller than that with the assumption. They can even become negative in some regions, a result that is not possible if liquid water content is held constant. However, this finding is consistent with results of model studies and other observations.

The cloud column susceptibility itself can be regarded as a description of the aerosol indirect effect. To this end, this study shows that the current aerosol indirect effect is largest over remote ocean areas (cf. Fig. 1b). As pollution increases, the aerosol indirect effect may become less and even saturated as evidenced by the small (sometimes negative) values of cloud column susceptibility over most continents and surrounding areas.

Retrievals of the cloud column susceptibility are based on the retrieval of cloud optical thickness and effective droplet radii. Due to uncertainties and possible bias involved in those retrievals, the cloud column number concentration was underestimated by 19% in comparison with *in situ* measurements [*Han et al.* 1998b]. However, this bias will not lead to a systematic bias for the cloud column susceptibility because in the retrieval of cloud column susceptibility, only the difference of N_c is used.

Currently, the results shown in this paper is calculated at 2.5°x2.5° degree grid boxes. We would be glad to calculate the cloud column susceptibility at different spatial resolutions for different GCM spatial resolutions.

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